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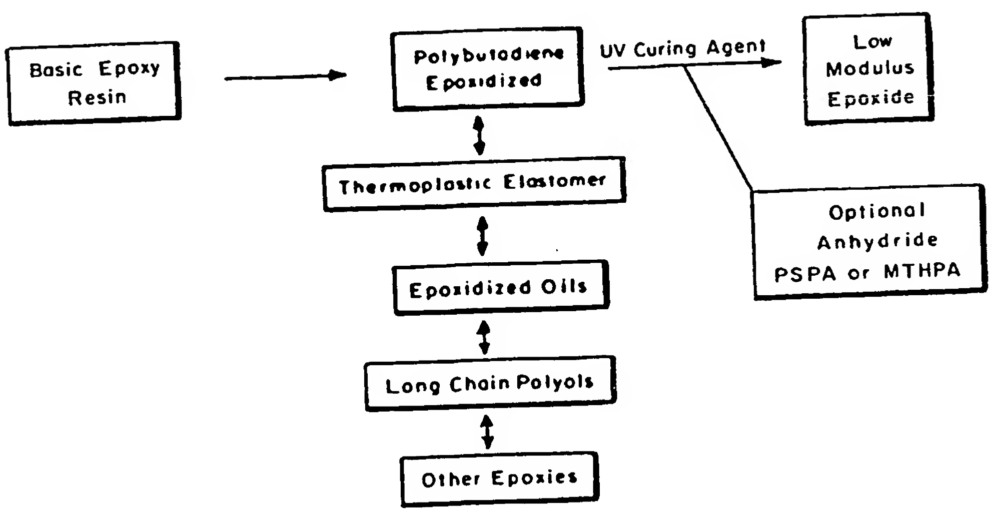

(57) Abstract

A UV curable composition comprising a UV curable epoxy-functional essentially solver: fee reactant, a UV curing agent. In certain embodiments, the composition also may comprise a conductive filler, which may be present at a selected concentration, particle size distribution and shape, wherein said composition, subsequent to cure, provides a deposited conductive film whose conductivity is controlled by said conductive filler concentration, particle size distribution and shape.

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UV-CURABLE EPOXY FORMULATIONS, INCLUDING CONDUCTIVE COMPOSITIONS

This application is a continuation-in-part of U.S. Patent Application Serial No. 09/104,601, filed June 24, 1998, incorporated herein by reference.

FIELD OF THE INVENTION

This invention is related to the use of conductive UV-curable epoxy formulations as coatings for substrates. More particularly, the formulations themselves are all UV-curable, and can be made as high elongation low moduli materials. In addition, the present invention relates to UV curable flame retardant materials, for use in the electronics industry, wherein such formulations can be made flame retardant or even self-extinguishing. The formulations herein also in alternative embodiment demonstrate excellent electrical insulation capabilities, and critically compensate for coefficient of thermal expansion mismatches between components in electronic board applications.

BACKGROUND OF THE INVENTION

Epoxy resins are generally characterized by the possession of more than one oxirane ring per molecule. The epoxy group itself may lie within the body of the molecule or in a terminal position. Epoxy groups are characterized as highly strained, and therefore, are quite reactive to many substances, particularly nucleophiles and proton donors. Such reactions allow chain extension and/or crosslinking to occur without the elimination of small molecule by-products. That being the case, epoxy formulations tend to exhibit a lower curing shrinkage than many other types of thermosetting plastics.

A very wide range of epoxy resins have been reported in the literature, most of which deal with the rigidity and high modulus behavior of these systems, in a highly crosslinked or thermoset configuration. In formulating such systems, the non-epoxy part of the system may be aliphatic, cycloaliphatic or highly aromatic hydrocarbon or it may

be non-hydrocarbon and quite polar. It may contain unsaturation. Similar remarks also apply to the chain extension/cross-linking agents, so that cross-linked products of great diversity may be obtained. In practice, however, the commercial scene has been dominated by the reaction products of bisphenol-A and epichlorohydrin which account for the majority of market share. For a short yet excellent review of epoxy chemistry, attention is directed to "Plastics Materials", by J.A. Brydson, Butterworths, 5^{th} Edition, 1989.

For a number of applications, however, epoxy resins may be considered to have a variety of disadvantages. These disadvantages include high viscosity, high cost, and too high a rigidity for specific applications. The resins therefore have been modified by incorporation of compounds known as dilutents and fillers, and for surface coating applications, blends with other resins.

More specifically, dilutents, by definition, are free flowing liquids incorporated to reduce viscosity and simplify handling. Typical dilutents, which arc also known as "reactive" dilutents, include phenyl glycidyl ether, butyl glycidyl ether and octylene oxide. Fillers include sand, metal powders, metal oxide fillers, wire wool and asbestos. Other additives include non-reactive types, fall into the general category of conventional phthalates and phosphates. Alternatively, polymeric additives have been employed, particularly low molecular weight polyamides from dimer acids, low molecular weight polysulphides, polyamines and the polyglycol diepoxides.

Specific examples of modified epoxy systems have also been widely reported in the patent literature, and the following are noted: 1. European Patent Application No. 9411 9996 describes a one component epoxy resin composition. The polyepoxide resins are said to comprise an epoxidized resin based on a polyglycidyl ether of a phenolic type compound, and a latent amine curative; 2. European Patent Application No.88810851, entitled "Flexibilized Epoxy Resin Compositions" describes an epoxy resin in

combination with a polyester which is carboxy terminated. The composition is described as useful as a surface coating or as an adhesive. In addition, heat curing or curing by means of combined treatment with actinic radiation and heat is disclosed; 3. European Patent Application No. 92810598 discloses a two component epoxy resin adhesive system composing an epoxy component containing at least one aromatic multi-functional epoxy resin and a liquid elastomer component comprising a liquid co-polymer based on butadiene and at least one ethylenically unsaturated comonomer and a liquid co-polymer reaction product of a polyamine and a dimer acid and a liquid aromatic or aliphatic polyamine. The compositions were said to have a high glass transition temperature, flexibility, and fast curing; 4. U.S. Patent No. 5,318,808 discloses compositions for photocurable coatings. The compositions are said to comprise (a) an epoxidized vegetable oil, (b) a low molecular weight epoxy resin, (c) a photoinitiator for cationic polymerization and (d) a wax. Processes for making and using and coatings arc also disclosed as are containers coated according to the invention; 6. U.S. Patent No. 5,516,824, entitled "Solvent-free Laminating Adhesive Composition," reports on a laminated composing an epoxidized block copolymer in combination what is identified as a 'tackifying" resin, compatible with the epoxidized block copolymer material. The epoxidized block copolymer is said to contain blocks of isoprene and butadiene, with varying molecular weight; 6. U.S. Patent No. 5,545,510, entitled "Photodefinable Dielectric Composition Useful in the Manufacture of Printed Circuits" defines a composition and process for fabricating circuitry packages. The composition is said to compose a carboxy functional resin, an acrylate oligomer, an epoxy functional resin, a butadicne nitrite resin, and a photoinitiator. The composition is claimed in connection with fabricating circuitry packages; 7. U.S. Patent No. 5,258,459, entitled "Resin Compositions for Coating and Electrodeposition Coating Composition Containing the Same", discloses a resin composition obtained by reacting an epoxy resin, a butadiene-acrylonitrile copolymer håving carboxyl groups or amino groups at both terminals of the molecule and (c) a bifunctional mononuclear phenolic compound, such as resorcinol. The coating compositions are described as having high adhesion to metal substrates, good

flexibility, chipping resistance and low temperature properties; 8. U.S. Patent No. 5,075,379, entitled "Rubber/Diamine Blends for Use in Curing Epoxy Resins," describes the use of a blend of diaminoisopropylbenzene and an amine terminated butadiene nitrite liquid rubber for curing epoxy resins. The blend is described as producing a rubber toughened epoxy resin having improved tensile properties; 8. U.S. Patent No. 4,482,660, entitled "Prepreg for Making A Composite having High Elongation . . .", discloses a prepreg formulated from an epoxy resin, the product of an epoxy resin and a butadiene-acrylonitrile copolymer having carboxyl groups on those terminals of the copolymer chain, and an amine curing agent. The composition produced is said to have tensile elongation and heat resistance; and 9. U.S. Patent No. 5,420,202 entitled "Epoxidized Low Viscosity Rubber Toughening Modifiers for Cycloaliphatic Epoxy Resins", describes a toughened cycloaliphatic epoxy resin comprising a curable cycloaliphatic epoxy resin, an epoxidized low viscosity polydiene polymer, and a curing agent.

The above review of basic epoxy formulations is relevant to the present invention, as the present invention relates to the use of high elongation and low moduli epoxy systems, in electronics applications, which has heretofore been unreported. However, before considering the details of the invention herein, it is worth noting that in general, electronic applications demand adhesive products, of which epoxies are excellent candidates, that possess a number of properties in addition to adhesive strength and compatibility with substrate surfaces. In this regard, attention is first directed to *Circuits & Assembly*, August 1995, wherein the inventor of this application points out that with respect to present theory and practice for surface-mount technology, including flip chip devices and ball grid arrays (BGAs), such theory and practice has emphasized the use of what is known as a rigid underfill encapsulant to maintain the viability of the device through temperature and humidity cycling.

Such stiff high modulus underfill materials are of course rigid. They are typically filled polymers (such as epoxies), and are designed to have a coefficient of thermal expansion (CTE) that approximates the other materials that they are in contact. Hence, during temperature cycling, it is expected that the encapsulant will expand and contract at a similar rate as the solder, ceramic, and chip, all of which have different CTE's making this approach difficult in reality. Due to the fact that the expansion is the same, little stress is generated on the complete package. However, a rigid underfill does not inherently possess a mechanism to dissipate stresses that occur during temperature and humidity cycling and/or mechanical vibration. That is, if the enapsulant is more rigid than the solder or other components, then stresses will be transferred into the solder bumps, joints and/or chips, potentially leading to failures.

Also, rigid organic polymers have different coefficient of thermal expansions (CTE's) above and below the Tg of the polymer. Thus, if the use or testing temperature of the devices varies through the Tg of the polymer, then stresses can be built up on the device by continually cycling through the Tg of the encapsulant. A constant rise in CTE through the use temperature range may ultimately prove to induce less stress on the system. In the final analysis, stress can be defined as: modulus multiplied by CTE. Different values of stress are therefore obtained below Tg and then from Tg to some elevated temperature.

What the above review therefore reveals is that with regards to underfill materials, and as part of the unique inventive concept herein, there has been a long-standing need for low moduli materials, to prevent damage during temperature cycling. More specifically, low modulus materials, in the case of surface mount technology, would provide a mechanism for mechanical and thermal shock to be dissipated. The desirable properties of such a systems would include, in addition to an extremely low modulus, superior elongation, low-moisture pickup and transport, minimal shrinkage upon cure,

rapid thermal cure, a CTE above that of the solder, a thermally conductive system, superior adhesion, and a Tg below or above device operating temperatures.

In addition, and with regards to flexible circuitry, there has been a similar long-standing demand for high elongation/low moduli type materials, as a replacement resin for various applications therein such that the flexible circuit design is properly protected by a suitable coating.

Towards this end, e.g., silicon adhesives have been proposed, and reference is made to the September/October 1996 issue of "Advanced Packaging", pages 30-31. Attention is also directed to *Advanced Packaging*, Winter 1993, and *The International Journal of Microcircuits and Electronics Packaging*, Vol. 19, No 3, Third Quarter 1996. Finally, attention is also directed to the IEEE Transactions on Components, Packaging and Manufacturing Technology, Part A, Vol. 17, No. 3, September 1994, which describes UV curable coatings for electronic components.

Furthermore, reference is made to the *Journal of Coatings Technology*, Vol. 66, No. 838, November 1994, which specifically reports on certain epoxidized polybutadiene as a reactive prepolymer for cationically curable photosensitive compositions. The certain coatings therein were said to posses high flexibility along with good adhesion to aluminum, steel and glass by adequate adjustment of the formulation compositions. However, this reference does not suggest and does not set moduli limits, as contemplated herein, nor the development of a low modulus system to deal with the above referenced problems of dissimilar CTEs in surface-mount electronic and coating systems and coating formulations.

All of the formulations noted above also have the disadvantage in that none of the resins therein collectively have the combined characteristics of being UV-curable along with a low moduli values, in addition to resistance to elevated temperature, good

electrical insulation capabilities, and superior adhesion, and as noted, the ability for the system to compensate for CTE mismatches in electronic board temperature cycling. In addition, none of the prior art resins are both UV curable and flame retardant regardless of modulus, or cure solely by UV treatment when highly filled with conductive material.

SUMMARY OF THE INVENTION

The present invention provides in a first aspect a UV curable composition comprising a UV curable epoxy-functional essentially solvent free reactant, and a UV curing agents said composition, subsequent to cure, exhibiting a modulus of less than about 50,000 psi, and an elongation of greater than about 3.0%.

In process form, the present invention describes a process for providing a low moduli underfill encapsulant which comprises the steps of mixing from 1 to 99 parts of a UV curable epoxy-functional essentially solvent free reactant with 99 to 1 parts of a component selected from the group consisting of epoxidized polybutadienes, thermoplastic elastomers, epoxidized oils, long chain polyols, other epoxy monomers/oligomers, and mixtures thereof, followed by addition of a UV curing agent and applying them to a substrate to provide a coated substrate and exposing said coated substrate to light of wavelength from about 100 to 700 nm wherein the mixture is substantially polymerized by exposure to said light, and said polymerized mixture has a modulus of less than about 50,000 psi and an elongation of greater than about 3.0 %.

In still further embodiment, the present invention also relates to a low moduli thermoset encapsulant which comprises an epoxy based thermoset resin with which dissipates stresses developed as between the encapsulant and other materials contacted due to differences in coefficient of thermal expansion of said other materials and said flexible thermoset encapsulant.

Furthermore, the present invention also relates to the preparation of a low moduli thermoset coating which comprises an epoxy based resin characterized in that the resin is UV curable, and has a moduli of less than about 50,000 psi, and an elongation of greater than about 3.0%. In addition,the coating can be made flame retardant or self-extinguishing, e.g., upon addition of a flame retardant additive selected from the group consisting of brominated compounds, phosphorous compounds, and antimony compounds.

The present invention also relates to novel UV curable epoxy based systems which contains a conductive material e.g. particulate filler thereby providing a conductive film subsequent to cure. Exemplary conductive additives include silver, copper, aluminum and the like.

Compositions of the invention also may contain a thermally conductive material, e.g. where the composition is employed as a heat sink in an electronic device or other article. For instance, thermally conductive compositions of the invention may include an additive of one or more of aluminum oxide, aluminum nitride and the like.

Compositions of the invention can be suitably applied on a variety of substrates. For instance, compositions of the invention can be applied on electro-magnetic shielding substrates; RF tag substrates; membrane switch substrates; interconnects for semiconductor devices and other microelectronic devices; electo-optical devices such as waveguides; printed circuit board substrates (e.g., as an innerlayer for multilayer boards) and other electronic packaging substrates.

Preferred compositions of the invention cure (e.g. crosslink) by an acid-promoted reaction, and without a free radical-based reaction. Hence, preferred compositions of the invention are substantially, essentially or completely free of a free radical initiator component, such as 2-hydroxy-2-methyl-1-phenyl-propan-1-one. More specifically,

preferred compositions of the invention contain less than about 5 or 2 weight percent based on curing agents of such free radical initiators, more preferably less than 1 or 0.5 weight percent of such free radical initiators, or still more preferably are completely free of such free radical initiators.

Preferred compositions of the invention also contain an acid generator compound to promote the curing reaction, and are substantially, essentially or completely free of any other agent to promote cure, such as an agent that promotes curing upon thermal treatment, i.e. a thermal curing agent. Such thermal curing agents that have been employed include amine complexes such amine complexes of BF_3. Preferred compositions of the invention contain less than about 5, 3 or 2 weight percent (based on total composition solids) of such thermal cure agents, more preferably less than 1 or 0.5 weight percent of such thermal curing agents, or still more preferably are completely free of such thermal curing agents.

References herein to modulus values (in psi, pounds per square inch) designate Young's modulus values as determined by ASTM D882 test protocols. References herein to elongation values designate values (expressed in percent increase of rupture or breakage length versus original length) as determined by ASTM D882 test protocols. Other aspects of the invention are discussed below.

BRIEF DESCRIPTON OF THE DRAWINGS

FIG. 1 illustrates in block diagram format the overall synthetic scheme and options therein for producing the UV curable low moduli epoxy formulation of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

The present invention in a first embodiment comprises a UV curable composition comprising a UV curable epoxy-functional essentially solvent free reactant, and a UV

curing agent; said composition, subsequent to cure, exhibiting a modulus of less than about 50,000 psi, and an elongation of greater than 3.0 %. Preferably the UV curable composition includes a component that is selected from the group consisting of epoxidized polybutadienes, thermoplastic elastomers, epoxidized oils, long chain polyols, other epoxy monomers/oligomers, and mixtures thereof.

Those skilled in the art will appreciate that by incorporating the above referenced components, the moduli values will be lowered, and elongational properties will be improved. In broad embodiment, therefore, the level of the UV curable epoxy-functional essentially solvent free reactant will vary between 1-99 % (wt) and the above referenced component will vary between 99-1%, in order to achieve the desired reduction in modulus and increase in elongational properties. Preferably, however, the UV curable reactant is present at a level of less than about 40% (wt), the thermoplastic elastomer is present at a level of less than about 5% (wt); the epoxidized oils are present at a level of less than about 40% (wt.); the long chain diol is present at a level of up to about 66% (wt) and the other epoxy monomers/oligomers are present at a level of up to about 50% (wt). In addition, the optional anhydride compound, if employed, is preferably present at a level of about 30-50% (wt.), which anhydride assists in the curing of the formulation.

Preferably, the UV curable epoxy-functional essentially solvent free reactant is formed by reaction of bisphenol A and epichlorohydrin and the UV curing agent generates acid in the presence of UV light. More preferably, the UV curing agent is an acid generating onium salt compound. In addition, and in optional embodiment the formulation may contain an anhydride functional compound such as polysebasic acid anhydride or methyl(tetrahydrophthalic anhydride). Such anhydrides also cure in the presence of UV light and the epoxide component and UV curing agent.

As indicated above, preferred compositions of the invention are essentially solvent-free. However, the compositions may contain relatively minor amounts of

solvent to facilitate coating, or to apply comparatively thinner composition coating layers. For example, compositions of the invention may suitably be formulated at about 50, 60, 70 or 80 or more weight percent solids, balance solvent, more typcially about 85, 90 or 95 or more weight percent solids, balance solvent. Preferred solvents include e.g. aromatic solvents such as toluene or xylene, or an esterified or etherified glycol such as propylene glycol methyl ether.

In process form, the present invention can be described as a process for providing a low moduli underfill encapsulant or an encapsulant, conformal coating which comprises the steps of mixing from 1 to 99 parts of a UV curable epoxy-functional essentially solvent free reactant with 99 to 1 parts of a component selected from the group consisting of epoxidized polybutadienes, thermoplastic elastomers, epoxidized oils, long chain polyols, other epoxy monomers oligomers, and mixtures thereof, followed by the addition of a UV curing agent, and applying the mixture to a substrate to provide a coated substrate and exposing the coated substrate to light of wavelength from about 100 to 700 nm wherein the mixture is substantially polymerized by exposure to said light. By such process, preferably, the modulus developed is less than about 50,000 psi. Preferred compositions of the invention exhibit even lower modulus values upon curing, e.g. a modulus of about 40,000 psi or less, about 30,000 psi or less, or even about 20,000 psi or about 10,000 psi or less.

In addition the present invention herein includes what can be termed a low modulus thermoset encapsulant which comprises an epoxy-based thermosetting resin with which dissipates stresses developed as between the encapsulant and other materials contacted due to differences in coefficient of thermal expansion of said other materials and said flexible thermoset encapsulant. Preferably, in such context, the thermoset encapsulant modulus is less than about 50,000 psi, or even lower as discussed above.

In analogous fashion, the present invention also provides a low moduli thermoset coating which comprises an epoxy based resin characterized in that the resin is UV curable, and has a moduli of less than about 50,000 psi, further characterized by an elongation of greater than about 3.0%. Preferred compositions of the invention exhibit even greater elongation values upon curing, e.g. an elongation value of about 5% or 10% or greater, more preferably about 20 or 30% or greater, or about 40%, 50%, 60%, 70%, 80%, 90% or 100% or more.

Table I below sets out the various representative formulations which have been found exemplary and preferable in accordance with the invention disclosed herein. For the formulations presented in Table 1, it is to be noted that the samples were all polymerized by a UV light source that was broad banded and centered around 365 nm. Table II lists the measured mechanical properties.

The following is representative of the epoxide formulations used.

The following materials were mixed (i.e. rolled) in a wide mouth jar for ½ hr: 40 grams of Epon 828 (Shell Chemical Company); 100 grams of polybutadiene (Elf Atochem); 100 grams Heloxy 505 (Shell Chemical Company). Then, 2.4 grams of CD 1010 (Sartomer Corp, an onium salt latent acid generator) was added and the reaction mixture was stirred for an additional 5 minutes.

Aerosil 200 (thixotropic silica gel) from Degussa was added as appropriate to thicken the same for obtaining a vararaible coating thickness. The samples were coated onto Mylar or release paper and polymerized by exposure to actinic radiation from a Dymax EC 5000 exposure tool with a broad-band radiation centered around 365 nm for 20 to 45 seconds.

Accordingly, the formulations which appear below in Table I were prepared according to the above general procedure, with the various amounts and optional reagents as indicated. Following Table I is Table II which presents the various mechanical properties (tensile strength, elongation at break, modulus and hardness values) of the formulations so prepared.

TABLE 1
FORMULATIONS

SAMPLE #	Heloxy 605	Heloxy 505	PS PA	Vicoflex 9010	828	Heloxy 48	Ricoturf	RP 30	KF-188	LD	HT 221
1	41.6	41.6			16.7						
2	41.6			41.6	16.7						
3						2.3	48.9	48.9			
4	50		16-		33.3						
5A	25		8.3		16.7				50		
5B	25		8.3		16.7				50		
6			20		40				40		
7	31.2	31.2			12.5				25		
8	20.3	20.3			8.15				51.2		
9	41.6				16.7						
10	34.7			34.7	13.9					11.9	
11	34.7			34.7	13.9					11.9	
12	34.7			34.7	13.9					11.9	
13	40	40			20						
14											100
15									26.8		73.2
16	16	16			8				50		
17	36.3	36.3	9.2		18.2						
18	25.01		8.3		16.66				50		
19	16.26	29.27			19.5				9.75		

TABLE II

PROPERTIES

SAMPLE #	T.S. (psi)	Elg. Break (%)	Mod. (psi)	Shore A
1	377	19.6	1926	85
2	626	30.1	2079	85
3	213	17.8	1196	70
4	720	17.95	4011	72
5A	52.5	61.0	86	55
5B	52.5	61.0	86	55
6	205	78.4	261	
7	137	21.5	637	
8				
9	264	19.5	1353	
10	443	22.4	1977	
11	343	24.4	1405	
12	184	18	1022	
13	144	8.4	1714	
14	287	3.7	7756	
15	335	7.5	4466	
16	4.9	36.7	13.3	
17	193	17.1	11.28	
18	134	28.2	47.5	
19	367	76.4	480	

As can be seen from Table I, the preferred epoxy resin is Epon 828, which is available from Shell, which is an epoxy resin formed from bisphenol A and epichlorohydrin and equivalents are available from Dow. It is to be noted that equivalents to such epoxy resins are available from Dow. In addition, other epoxy resins which have been employed include Heloxy 505, Heloxy 84, and Heloxy 48, which are also epoxy resins (glycidyl ethers of castor oil), and again, available form Shell. Limonene dioxide (LD) is also employed, available from Elf Atochem, which is an aliphatic epoxy acting to dissolve the thermoplastic elastomer. Vicoflex 901 0 is an epoxidized lincensed oil/methyl ester. Ricotuff 11100/A is a maleated polybutadiene. KF-

188 is a polyol. LD is reference to limonene dioxide. HT-221 is High Temp 220, a commercially available epoxy resin.

The preferred reactive polybutadienes include hydroxy terminated epoxidized polybutadienes, typical of which are Poly BD 600 and Poly BD 605 available from Elf Atochem. However, in the broad scope of the invention herein, it will be appreciated by those skilled in the art that the hydroxy termination can be supplied sources other than a polybutadiene product. For example, such alternative sources would include a hydroxy terminated polyester. In addition, the invention herein in preferred embodiment makes use of Ricotuff 1100/A. Such maleated polybutadiene was found particularly useful in further improving elongation and lowering of the modulus.

Other reactive components include anhydrides including polyanhydrides e.g. polysebasic polyanhydride, available from Lonza as well as methyltetrahydrophthalic anahydride. Such components act to increase elongation and decrease modulus and improve thermal stability. Furthermore, a liquid polyisoprene polymer can be incorporated herein, at levels of about 1-10%, and said liquid polyisoprene also acts to reduce moduli values.

With regard to the optional use of a thermoplastic elastomer as a further component to the formulation described herein, it has been found preferable to employ Europrene SOL T 190 or SOL T-166 or KRATON FG which are all styrene based thermoplastic elastomers, all of which are particularly useful in for increasing elongational properties.

Finally, in all of the formulations, it is possible to employ a small amount of an adhesion promoter (about 1%), and in this regard, Dynasylan Glymo has been found as a preferred promoter, available from Huls. If required, an adhesion promoter can be added to a composition of the invention.

In addition, the preferred UV curable acid-generating curing agent was an opium salt, preferred of which include 1:2 mixtures of Sartomer 1012/LD or Sartomer 1010 or Sartomer 1011 or equivalents available from Union Carbide. A variety of other onium salts also may be employed as the UV-activated acid generator, including sulfonium and iodonium compounds , such as compounds disclosed in European Published application 0783136. See also U.S. Patent 5731364 to Sinta et al. for additional useful sulfonium compounds.

Other photoacid generators compounds can be employed in compositions of the invention, either in place of or in combination with an onium salt. For instance, imide photoacid generator compounds can be employed such as imidosulfoantes and N-sulfonyloxyimides. See International application WO94/10608 exemplary imide acid generators. Nitrobenzyl photoacid generator compounds also may be employed, such as those nitrobenzyl photoacid generators disclosed in European Published application EP717319A1. Disulfone photoacid generators also can be employed, such as the compounds disclosed in European Published application 708368A1.

Furthermore, while the above formulations provide suitable properties for a low moduli coating application, it has been found as a preferred alternative that in some cases, post curing, via thermal curing techniques, is desirable. That is, in certain systems where thickness becomes significant, both UV and thermal cure can be preferably employed. With respect to such dual curing, those skilled in the art will appreciate that in the case of such thick coatings, UV curing may only be relatively efficient at or near the surface, and in such cases, a conventional thermal curing process can be applied to insure total cure throughout the cross-section of such thick coating formulations. Suitable post-UV exposure thermal treatment may include heating at 60°C, 70°C, 80°C or 100°C for 0.5 to 2 minutes.

Compositions of the invention also may be exposed to patterned UV light such as through a photomask to provided a patterned image in a composition coating layer. The patterned compositions then may be developed (either by wet or dry treatments) to remove coating areas not exposed to the activating radiation, i.e. to provide a negative relief image. Suitable wet developers can vary with the composition components and can include an aqueous alkali solution, or an organic solvent developer, as is generally known in the art. Dry development with a plasma also may be employed. Such use of compositions of the invention as a negative-acting resist can beneficial in many applications, e.g. where a highly precise coating layer is required, such as in manufacture of a semiconductor or other electronic device.

With reference to Table II above, it can be seen therein that a true low moduli UV curable epoxy formulation has been developed, with associated elongational values. As shown therein, by formulating according to Table I, modulus values as low as 47.5 psi, and upwards of 8000 psi, have been obtained. In addition, elongations of at least about 3.0 % are herein obtained Furthermore, those skilled in the art will appreciate that by adjusting the formulation, moduli values can be increased. That is, in accordance with the basic concept herein, of developing an essentially solvent free UV curable epoxy resin, moduli values can be increased incrementally to 10,000 psi, 20,000 psi, 30,000 psi, 40,000 psi, 50,000 psi, and up, until the appropriate moduli is obtained. Towards such end, one increases the amount of UV curable epoxy resin relative to, e.g., the epoxidized polybutadienes, thermoplastic elastomers, epoxidized oils, long chain polyols, and other epoxy monomers/oligomers, while maintaining UV curing capability.

Attention is directed to FIG. 1, which illustrates the basic overall approach for preparing the low moduli epoxy systems of the present invention. As illustrated therein, the basic epoxy resin can be combined with, e.g. epoxidized polybutadicnes, thermoplastic elastomers, epoxidized oils, long chain polyols, and/or other epoxy monomers/oligomers, or mixtures thereof. Accordingly, those skilled in the art will

recognize that one aspect of the invention herein is to select a component which will, together with the UV curable epoxy-functional essentially solvent free reactant, provide the low moduli materials disclosed herein. The amount of such component, as previously noted, has been found to vary as between 1-99 parts, relative to 99-1 part of the basic epoxy reactant. Also illustrated in FIG. 1 is that a UV curing agent is included, and optionally, an anhydride functional compound which provides, in addition to the epoxy functionality, another chemical moiety for curing.

With regard to the utility of the present invention, the low moduli formulations herein, apart from utility for dealing with CTE mismatches between components and the board, will also have broad utility in armature and cartridge attachment, buffer coats, chip bonding, chip on board, coil bonding, conformal coatings, glob tops, high flow IC cards, hybrid module potting, interlevel dielectrics, LED coatings, magnetic and steppermotor assemblies, optical potting, opto electronics, smart cards, and wire tacking.

In addition, in preferred embodiment, the formulations herein are UV curable low moduli epoxy/polybutadiene combinations, whereas current UV curable systems, as used in the electronic industry, depend upon acrylates, urethanes and silicons. These latter materials are more polar, and hence, the epoxy/polybutadiene formulations herein have better electrical properties and lower moisture absorption than these prior art systems. That is, preferred compositions of the invention are substantially, essentially or completely free of acrylate, urethane and/or silicon components. For example, preferred compositions of the invention contain less than about 10, 5, 3 or 2 weight percent (based onototal composition solids) of an acrylate, urethane and/or silicon component, more preferably less than about 1 or 0.5 weight percent of an acrylate, urethane and/or silicon component, or even more preferably the composition is completely free of an acrylate, urethane and/or silicon component.

Furthermore, polymerization of epoxides (as occurs upon curing of compositions of the invention) is a ring opening mechanism and therefore there will be less shrinkage upon cure than with acrylate products. Furthermore the epoxies themselves after crosslinking are resistant to solvents, and the products formulated herein can be dual curable, which allow for thermal post curing.

In addition, as noted in the summary of the invention, also disclosed herein is a UV curable composition which comprises an epoxy-functional essentially solvent free epoxy resin which is UV curable, which therefore contains a UV curing agent, wherein such composition, subsequent to cure, is substantially flame retardant or self-extinguishing. The composition can have an added component to impart flame resistance, or a primary component of the composition can be modified to impart flame resistance, in particular a halogenated epoxy material can be employed, preferably a brominated epoxy material. A halogenated material such as a halogenated epoxy component can be sufficient to impart acceptable flame retardant properties.

Below is an example of a preparation that illustrates the preparation of such flame retardant UV curable epoxy based formulation.

120 grams of Epox 1163 (a brominated bisphenol A) from Shell is melted in an oven at 125 C. To the warm solution was added 120 grams of Heloxy 505 and 50 grams of PHT-4 diol (3,4,5,6-tetrabromo-1, 2-benzene dicarboxylic acid; mixed esters with diethylene glycol and propylene glycol) available from Great Lakes Chemical, and stirred until a homogeneous solution was obtained. To 50 grams of said solution was added an additional 5 grams of PHT-4 diol.

In a separate vessel 90 grams of Epoxy 828, 180 grams of Polybutadiene 605 and 120 grams of Heloxy 505 were mixed. 10 grams of this solution were added to the above solution.

To the above mixture (65 grams) was added 2.68 grams of TNPP (trinonylphenyl phosphine) (General Electric Specialty Chemicals) followed by 790 mg of Sartomer 1010 (cationic onium antimony salt). The mixture was then stirred until homogenous. To the rapidly stirring mixture of the above, 2.25 grams of antimony trioxide (United Mineral & Chemical Corp.) and 670 mg of Aerosil 200 (Degussa) was added and mixed vigorously for 15 minutes. The formulation was then degassed under vacuum and coated onto substrates and cured into a film by a 45 second exposure.

A nominal 5 mil coating was made on Mylar and release paper. After cure, when the product was removed from the release paper and held above a flame it did not ignite and self-extinguished. The same coated onto Mylar was exposed to actinic radiation on both sides of the film for a total of 90 seconds (45 seconds on each side). When placed in a flame the sample was self-extinguishing. As referred to herein, the terms "substantially self-extinguishing" or "substantially fire retardant" indicates that the composition does not ignite under the above described test of a 5 mil coating of the composition being placed in a flame.

A wooden tongue depressor was dipped into the above formulation. The coating was cured by actinic radiation. When placed in a flame the wooden tongue depressor failed to ignite.

As noted above, in further embodiment the present invention also relates to novel UV curable epoxy based systems composing a UV curable epoxy-functional essentially solvent free reactant, and a UV curing agent, and a conductive filler, wherein said conductive filler is present at a selected concentration, particle size distribution and shape, and wherein said composition, subsequent to cure, provides a deposited conductive film whose conductivity is controlled by said conductive filler concentration,

particle size distribution and shape. In addition, it has also been determined that the type of UV curable epoxy based system can also effect conductivity.

Accordingly, it has been determined that the various low moduli UV curable systems notes above, as well as various other UV curable epoxy systems with higher moduli values, can be combined with conductive particular fillers as noted above to provide resistance as low as the milliohm range and up to several kiloohms as described in the following working examples which appear below. More specifically, resistance of about 500 milliohms or less can be achieved in the conductive films herein. Suitable but by no means limiting conductive fillers preferably include silver or silver flake, copper, aluminum, solder powder, and mixtures thereof. A particularly preferred conductive filler is a silver/solder mixture. Preferred loading of conductive filler is, as noted, controlled to effect conductivity, and in preferred embodiment is about 50% (wt.) and higher, based on weight of total composition solids. However, other loading levels also will be suitable, e.g. compositions that contain condutive material at a concentration of about 10 wt. % or greater, based on total composition solids, or at a concentration of about 20 wt% or greater, 30 wt. % or greater, 40 wt. % or greater, 50 wt. % or greater, 60 wt. % or greater, 70 wt. % or greater, oe even 80 or 85 wt. % or greater, based on total solids of the composition. The following illustrative example is provided:

BASE-POLYMER PREPARATION

PRODUCT	WT. (GMS)	COMPANY
PBD 605	20.3	Elf Atochem
Heloxy 505	20.3	Shell
Epon 828	8.15	Shell
K-Flex 188	51.2	King Industries
CD1010	0.752	Sartomer

- Product 1: 4.0 gms Ag flake (Technic 556)/1.25 gms Base Polymer Preparation
- Product 2: 3.0 gms Ag flake (Technic 556)/3.0 gms 6337 solder powder (Kester, mesh size, filtered (meshed) or non-meshed 325-500)/1.87 gms Base Polymer Preparation.

Mix each of the above by hand. Draw down using draw down bar set at 3-5 mils. Coatings surface either paper or Kapton® (Dupont). Before use the Kapton® was heated at 150°C for 5 minutes to remove the moisture from the film.

Samples were cured by a 400 watt metal halide lamp system centered at 365 nm (**Dynmax** EC 5000 - 675.9 hrs on the bulb). Cure cycle was either:

- 45 secs. Exposure - 1 minute hold - 45 secs. Exposure (A)
- 45 secs. Exposure (B)

The resistance of the samples was measured using a Keithly model 580 microhm meter using a spring-loaded tip probe.

After UV cure generally a 2-3 mil thick film was obtained: Results are shown below:

Product	Sample #	Substrate	Exposure	Thickness	Resistance (milliohms)
1	161-3A	Kapton	A	2	7.9
1	161-3B	Paper	A	2	4.5
1	161-4	Paper	B	2	4
2	161-1	Paper	B	2	11-15
2	162-2	Kapton	B	2	26

All documents mentioned herein are incorporated herein by reference.

The foregoing description of the invention is merely illustrative thereof, and it is understood that variations and modification can be made without departing from the spirit or scope of the invention as set forth in the following claims.

I claim:

1. A UV curable composition comprising a UV curable epoxy-functional essentially solvent free reactant, and a UV curing agent, the composition, subsequent to cure, exhibiting a modulus of less than about 50,000 psi, and an elongation of greater than about 3.0%.

2. A UV curable composition comprising a UV curable epoxy-functional essentially solvent free reactant and a UV curing agent, and a conductive filler, said conductive filler present at a selected concentration, particle size distribution and shape, wherein said composition, subsequent to cure, provides a deposited conductive film whose conductivity is controlled by said conductive filler concentration, particle size distribution and shape.

3. A UV curable composition comprising a UV curable epoxy-functional essentially solvent free reactant and a UV curing agent, and a conductive filler.

4. The composition of claim 2 or 3 wherein salt conductivity is measured by a resistance of less than about 500 milliohms as measured in said film as deposited.

5. The composition of claim 2 or 3 wherein said resistance is less than about 25 milliohms.

6. The composition of claim 2 or 3 wherein said conductive filler comprises silver, copper, aluminum, solder powder and mixtures thereof.

7. The composition of claim 2 or 3 wherein said conductive filler comprises a silver/solder mixture.

8. The composition of claim 2 or 3 wherein said conductive filler is present at a concentration of about 50% (wt) or higher.

9. The composition of any one of claims 1-8 wherein the composition comprises an epoxy monomer or oligomer.

10. The composition of any one claims 1-8 wherein the composition comprises a component that is selected from the group consisting of epoxidized polybutadienses, thermoplastic elastomers, epoxidized oils, long chain polyols, and mixtures thereof.

11. The composition of any one of claims 1-8 wherein the epoxy-functional reactant is formed by reaction of bisphenol A and epichlorohydrin.

12. The composition of any one of claims 1-11 wherein the UV curing agent generates acid in the presence of UV light.

13. The composition of any one of claims 1-12 wherein the UV curing agent is an onium salt.

14. The composition of any one of claims 1-13 further comprising an anhydride functional compound in an amount of 30-50 weight percent.

15. The composition of claim 14 wherein the anhydride compound is polysebasic acid anhydride or methyl(tetrahydrophthalic anhydride).

16. The composition of claim 1 wherein the moduli is less than about 40,000 psi, preferably less than about 30,000 psi, more preferably less than about 20,000 psi, still more preferably less than about 10,000 psi.

17. The composition of any one of claims 1-16 wherein the elongation is about 10% or greater, more preferably about 20% or greater, still more preferably about 30% or greater, even more preferably about 50% or greater.

18. A process for providing a low moduli underill encapsulant which comprises

mixing from 1 to 99 parts of a UV curable epoxy-functional essentially solvent-free reactant with 99 to 1 parts of a component selected from the group consisting of epoxidized polybutadienes, themoplastic elastomers, epoxidized oils, long chain polyols, other epoxy polymers, and mixtures thereof;

adding a UV curing agent;

applying the mixture to a substrate to provide a coated substrate and exposing the coated substrate to light of a wavelength from about 100 to 700 nm wherein the mixture is substantially polymerized by exposure to the light, and thermally post-cured.

19. The process of claim 18 wherein the modulus of the polymerized mixture is less than about 50,000 psi.

20. The process of claim 18 wherein the composition comprises an acid generator compound that is activated by UV radiation, preferably an onium salt compound.

21. A thermoset encapsulant/coating which comprises an epoxy based thermoset resin cured by UV light which dissipates stress developed as between the encapsulatn and other materials contacted due to differences in coefficient of thermal expansion of the other materials and the flexible thermoset encapsulant, wherein the encapsulant/coating has a moduli of less than 50,000 psi and an elongation of greater than 3.0%.

22. The thermoset encapsulant of claim 21 wherein the moduli is less than about 40,000 psi, preferably less than about 30,000 psi, more preferably less than about 20,000 psi, still more preferably less than about 10,000 psi.

23. The thermoset encapsulant of claim 21 the elongation is about 10% or greater, more preferably about 20% or greater, still more preferably about 30% or greater, even more preferably about 50% or greater.

24. A low moduli thermoset coating/encapsulant which comprises an epoxy based resin that is UV curable and has a moduli of less than about 50,000 psi and an elongation of greater than about 3.0%.

25. A UV curable composition comprising a UV curable epoxy-functional essentially solvent-free reactant, and a UV curing agent, including a flame retardant component.

26. The composition of claim 25 wherein the composition comprises a brominated compound, phosphorous compound, antimony compound, or mixtures thereof.

27. The composition of claim 25 wherein the epoxy reactant is brominated.

28. The composition of any one of claims 25-27 wherein the composition further comprises a polyol compound.

29. The composition of claim 28 wherein the polyol compound is halogenated, preferably brominated.

30. The composition of claim 25 wherein the composition, subsequent to cure, exhibits a modulus of less than about 50,000 psi and has an elongation of greater than about 3.0 %.

31. A composition of any one of claims 25-31 wherein the composition is substantially flame retardant or substantially self-extinguishing.

32. A substantially flame retardant UV curable composition comprising a UV curable epoxy-functional essentially solvent-free reactant, and a UV curing agent.

33. The composition of claim 32 wherein the epoxy reactant is brominated.

34. The composition of any one of claims 32-33 wherein the composition further comprises a polyol compound.

35. The composition of claim 34 wherein the polyol compound is halogenated, preferably brominated.

36. An article of manufacture comprising a substrate having thereon a composition of any one of claims 1-17 or 21-35.

37. An article of claim 36 wherein the substrate is an electro-magnetic shielding substrate, RF tag substrate, membrane switch substrate, microlectronic wafer substrate, electro-optical device substrate, or a printed circuit board substrate.

38. An article of claim 36 or 37 wherein a coating layer of the composition on the substrate has been cured by exposure to activating radiation.

39. A circuit board substrate having coated thereon a composition of any one of claims 1-17 or 21-35.

40. A method for forming a composition relief image, comprising:

applying a coating layer of a composition of any one of claims 1-17 or 21-35 onto a substrate;

exposing the coating to patterned activating radiation and developing the exposed coating layer to provide a relief image of the coating layer.

41. The method of claims 40 wherein the substrate is an electro-magnetic shielding substrate, RF tag substrate, membrane switch substrate, microlectronic wafer substrate, electro-optical device substrate, or a printed circuit board substrate.

42. The method of claim 41 wherein the relief image is a cross-linked negative-tone image of the coating layer.

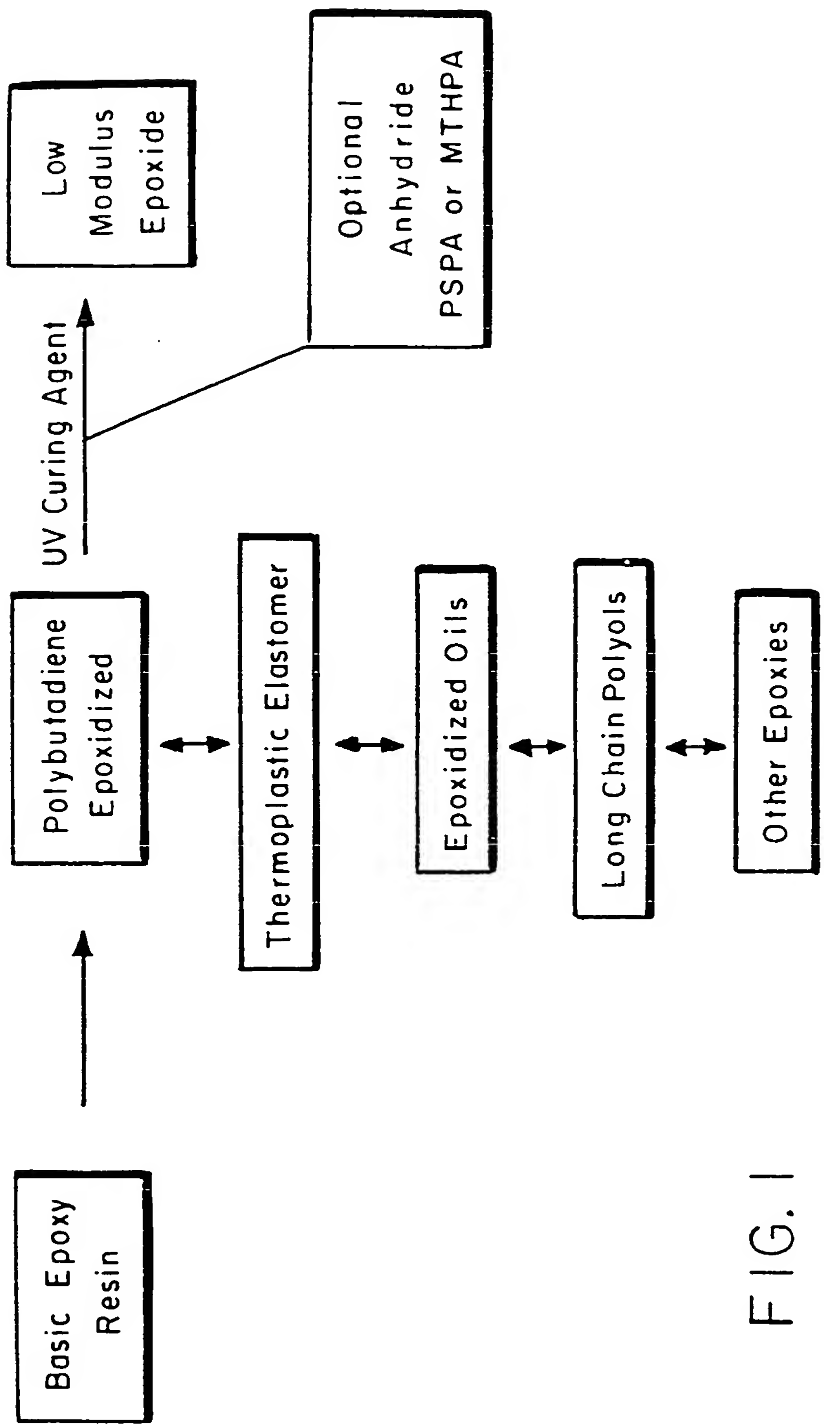
Basic Epoxy Resin
Polybutadiene Epoxidized
UV Curing Agent
Low Modulus Epoxide
Optional Anhydride PSPA or MTHPA
Thermoplastic Elastomer
Epoxidized Oils
Long Chain Polyols
Other Epoxies

FIG. 1

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US99/14531

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :H01B 1/02; CO8G 59/22; C08K 3/08
US CL : 522/100,170,181,81,79,83,71,25,31; 252/511,518,501.1,514,513
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 522/100,170,181,81,79,83,71,25,31; 252/511,518,501.1,514,513

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

Please See Extra Sheet.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X ---- Y	US 5,155,143 A (KOLESKE) 13 October 1992 (13.10.92), entire document.	1, 11-13, 16-24,28,,36-39 ----------- 2-8,14-15, 25-27,29-3 5,40-42
X	US 5,514,729 A (DIAMANT ET AL) 07 May 1996 (07.05.96), see entire document.	1-8,11-13, 16-25, 28, 36-42
X	US 4,999,136 A (SU ET AL) 12 March 1991 (12.03.91), see entire document.	1-8, 11, 16-17, 21-24, 36-42
A	US 5,667,899 A (YUSA ET AL) 16 September 1997 (16.09.97), see entire document.	1-42

[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
24 AUGUST 1999	27 OCT 1999

Name and mailing address of the ISA/US	Authorized officer
Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	SANZA MCCLENDON
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C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5,717,054 A (SCHULTZ) 10 Feburary 1998 (10.02.98),see entire document.	1-42
A	US 5,783,713 A (SCHULTZ ET AL) 21 July 1998 (21.07.98), see entire document.	1-42
A	US 5,703,195 A (SCHULTZ ET AL) 30 December 1997 (30.12.97), see entire document.	1-42
Y	US 3,983,075 A (MARSHALL ET AL) 28 September 1976 (28.09.76), see entire document.	1-6,8-9,11,14-17, 21-25, 36
Y	US 4,554,094 A (BABLER ET AL) 19 November 1985 (19.11.85), see entire document.	1-6,8-10,14 ,16-25,36-39
Y	US 5,589,714 A (HOWARD) 31 December 1996 (31.12.96), see entire document.	1-14, 16-28, 30-34, 36-42
Y	US 5,627,107 A (HOWARD) 06 May 1997 (06.05.97), see entire document.	1-14,16-28,30-34,36-42
A	US 5,700,581 A (SACHDEV ET AL) 23 December 1997 (23.12.97), see entire document.	1-42
A	US 4,435,214 A (EHRREICH) 06 March 1984 (06.03.84), see entire document.	1-42

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US99/14531

B. FIELDS SEARCHED
Electronic data bases consulted (Name of data base and where practicable terms used):

WEST 1.1A
SEARCH TERMS: CONDUCTIVE FILLERS, EPOXY, EPOXIDE,POLYOL, ANHYDRIDE, ONIUM SALT, UV CURING, PHOTOINITIATOR, UV RADIATION, FLAME RETARDANT, BROMINATED EPOXY, HALOGENATED EPOXY, HALOGENATED POLYOL,